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Coal-CO_2 Slurry Feed for Pressurized Gasifiers: Slurry Preparation System Characterization and Economics

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Abstract

Gasification-based plants with coal-CO_2 slurry feed are predicted to be more efficient than those with coal-water slurry feed. This is particularly true for high moisture, low rank coal such as lignite. Nevertheless, preparation of the CO_2 slurry is challenging and the losses associated with this process have not been accounted for in previous analyses. This work introduces the Phase Inversion-based Coal-CO_2 Slurry (PHICCOS) feeding system, in which coal-CO_2 slurry is prepared at ambient temperature *via* coal-water slurry. Steady-state process simulation is used to estimate the performance of the proposed slurry preparation and feeding system for bituminous coal and lignite. An Integrated Gasification Combined Cycle (IGCC) power plant with carbon capture is used here as a potential application, but this concept is applicable to any high-pressure coal feeding process. The economic attractiveness of the PHICCOS feeding system is assessed through calculation of its capital costs and resulting levelized cost of electricity, relative to competing commercial technologies. The findings of this work show that the PHICCOS feeding system offers a good tradeoff between overall process performance and costs. It is the most cost-effective method for feeding lignite and the second most attractive for bituminous coal, for which the competing technology is marginally cheaper. The PHICCOS feeding system is hence the only feeding system which is consistently cost-effective across the entire coal rank spectrum and is increasingly so for high-moisture and high-ash coal.

Keywords: coal; CO_2; slurry; gasification; feed; economics; ash

1. Introduction

The conveying of coal into pressurized gasifiers through slurry preparation and pumping is an attractive alternative to dry feeding based on lock hoppers. Slurry feeding is simpler and cheaper, can achieve higher pressures, and does not require feedstock drying. Nonetheless, systems based on coal-water slurry feed suffer from low thermal efficiency. This is a result of the large amount of energy used in heating up and vaporizing the slurry water, which is especially problematic for high-moisture coal.

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Liquid carbon dioxide has been suggested as an alternative to water for the preparation of coal slurry in plants with carbon capture [1-4]. An up to 12%-point higher gasifier cold gas efficiency has been estimated for coal-CO_2 slurry-fed systems as a result of the lower heat capacity and enthalpy of vaporization of CO_2, relative to water [4].

Unlike coal-water slurry, however, coal-CO_2 slurry cannot be prepared at ambient pressure: the triple point pressure of CO_2 is 5 bar so CO_2 cannot exist in its liquid state at lower pressures including ambient. Various methods for preparing coal-CO_2 slurry have been suggested, all of which use lock hoppers to overcome the minimum pressure required to form $CO_{2(l)}$, whereas chilling is often proposed as a way to reduce the minimum lock hopper pressure [5-7].

This work presents and evaluates an alternative approach, which allows for the preparation of coal-CO_2 slurry at ambient temperature and without the use of a lock hopper system. The Phase Inversion-based Coal-CO_2 Slurry (PHICCOS) preparation and feed proposed here is partly based on the Liquid Carbon Dioxide (LICADO) process, developed in the 1980's for the removal of inorganic sulfur from pulverized coal.

The main characteristics of the LICADO process for coal beneficiation are presented first, together with a brief description of the process development work conducted at the time as well as of its main findings. Next, the PHICCOS process for the preparation of coal-CO_2 slurry as a gasifier feedstock is introduced. Its performance is estimated based on steady-state process simulation relying on experimental observations for the LICADO process. Finally, a first estimate of the costs of the PHICCOS process is presented and compared with other feeding systems to arrive at an overall assessment of the attractiveness of this slurry preparation method and of its potential application for low and high-rank coal gasification. An Integrated Gasification Combined Cycle (IGCC) power plant with carbon capture is used in this work as an example application to quantify the merits of the PHICCOS process.

2. The LICADO and PHICCOS processes

The selective agglomeration of fine coal is a long-known process originally developed for coal beneficiation. Hereby, a water-immiscible liquid is mixed with coal-water slurry with the purpose of separating the inorganic impurities from coal. The process is based on the preferential wetting of the hydrophobic coal surface by the non-aqueous medium and the preferential wetting of the mineral impurities by water. The non-aqueous phase displaces water from the coal surface through adsorption onto its carbonaceous components, a process known as phase inversion [8, 9]. The coal-rich non-aqueous phase can be separated from the mineral-rich aqueous phase.

Several coal cleaning processes based on selective agglomeration have been developed and some have been commercialized. Nonetheless, the high operating costs related to the consumption and recovery of the non-aqueous medium, also known as agglomerant, have not allowed the process to be economically attractive in the long run. Fuel oil has been traditionally used as the agglomerant, however, other liquids such as n-pentane, n-heptane, and liquid carbon dioxide have also been considered [8,10].

2.1. The LICADO process for coal desulfurization

The LICADO process was developed in the 1980's for the purpose of coal beneficiation and desulfurization. It is a selective agglomeration process operating at 21°C and 60 bar. Liquid carbon dioxide is used both as an agglomerating agent and as a transport medium for the low-ash coal product. The separation principle is schematically illustrated in Figure 1.

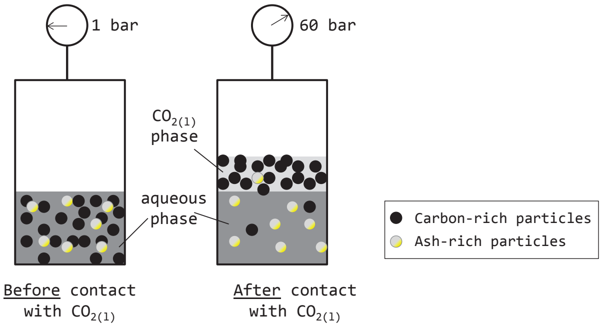

Figure 1: Separation principle of LICADO process (adapted from [11])

The LICADO process was developed by Westinghouse and the University of Pittsburgh with funding from the U.S. Department of Energy (DOE). The feasibility of this beneficiation method was first tested in a batch research unit with a volume of 3 L; a 6 L research development unit operating in semi-continuous mode followed. A fully automated continuous research unit with a capacity of 10 lb/h of beneficiated coal was then built and successfully operated with the purpose of gathering data for the detailed design of a 1 ton/day pilot unit. The experiments were conducted on eight different bituminous coals [12].

The LICADO experience showed that a coal particle size of 200 mesh (74 µm) is a good compromise between beneficiation efficiency and grinding energy. A very low product coal ash content of 2-5% (dry basis) was achieved consistently, even for coal with a high as-received (ar) ash content of 27%. Nonetheless, some of the coal is entrained in the aqueous stream, resulting in an enthalpy recovery of 85-90%. A low moisture content of 5-10% was measured consistently for the clean coal product from all bituminous coals studied[†]; it is a consequence of the displacement of water by liquid CO_2 and has been identified as one of the main advantages of the LICADO process, relative to other coal beneficiation methods producing a very wet product.

The LICADO process was designed to maintain CO_2 in its liquid phase in order to minimize CO_2 recompression costs. This, nonetheless, comes at the expense of high-pressure equipment, such as a pressurized auger filter for clean coal separation and lock hoppers for coal-water slurry preparation. These have been identified as the main reason for the high costs associated with this technology [8].

Interest in coal desulfurization ended, and with it the LICADO development, before the process operating conditions could be optimized and the pilot-scale plant was never built. Nevertheless, the results from the three experimental units provide sufficient data to indicate the expected process performance as well as the sensitivity of the process to specific design and operating variables such as particle size, agitation speed, $CO_{2(l)}$ flow, residence time, etc. A detailed engineering design of the individual pilot plant components was finalized based on these results, together with a budgetary estimate of the capital and operating costs of a commercial-scale unit producing 200 ton/day of beneficiated coal.

2.2. Phase Inversion-based Coal-CO_2 Slurry preparation: the PHICCOS process

The proposed PHICCOS process for the preparation of coal-CO_2 slurry at ambient temperature is presented in Figure 2. The CO_2 slurry is prepared via coal-water slurry (CWS); the phase inversion behavior, observed in the LICADO experiments, allows the separation of coal from the aqueous phase in the presence of CO_2.

[†] The as-received coal moisture of the bituminous coals is not reported in the original work [12]

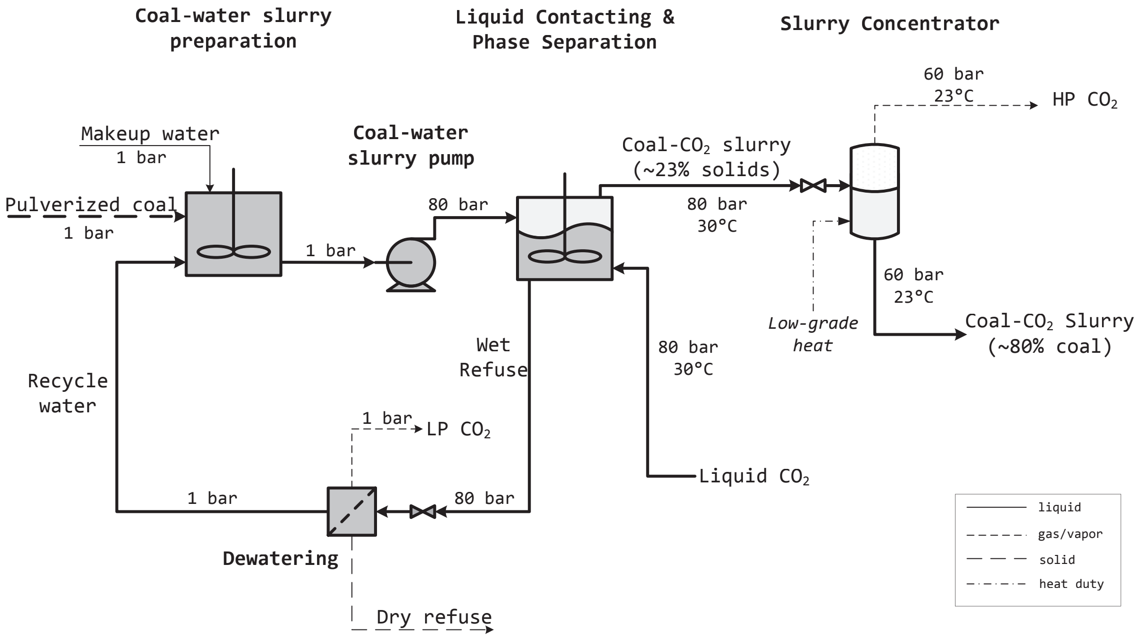

Figure 2: Schematic of proposed PHICCOS preparation and feeding system

Coal-water slurry is prepared in a conventional slurry preparation unit at ambient conditions. It is pumped to the same pressure as that of the liquid CO_2 stream available for slurry preparation. The term liquid CO_2 is used to refer to CO_2 with a liquid-like density, even though the pressure may be above the critical point.

The coal-water slurry is thoroughly mixed with liquid carbon dioxide, forming a water-rich and a CO_2-rich phase. Exposure of the coal surface to CO_2 leads to phase inversion; the low-ash, hydrophobic coal particles thus accumulate in the lighter CO_2 phase whereas high-ash, hydrophilic particles and moisture remain in the denser, aqueous phase. The two phases can be continuously removed from the top and bottom of the mixing vessel, respectively.

The aqueous ash-rich refuse leaving the liquid contacting vessel is brought to ambient pressure and dewatered before separating and disposing the high-ash solid stream. The refuse water is recirculated back to the coal-water slurry mixing vessel. Low-pressure (LP) CO_2 desorbed from the refuse during decompression is recompressed in the plant's CO_2 compressor.

The CO_2 slurry separated from the top of the mixing vessel has a low coal loading of about 20-25 weight-% (wt.-%), which, if fed directly to the gasifier, results in an excessively high CO_2 recirculation in the system. The coal-CO_2 slurry is hence concentrated before it is fed to the gasifier. Its CO_2 content is reduced to achieve a coal loading of 80% [4]. This is achieved by evaporating the excess CO_2 in the slurry concentrator through a combination of pressure reduction and low-grade heat addition. The vaporized, high-pressure (HP) CO_2 is recompressed and the concentrated, pressurized coal-CO_2 slurry is fed to the gasifier.

While the liquid contacting and separating unit of the PHICCOS process uses the same principle as the LICADO process, the overall process design and target application are different. Unlike the LICADO process, for example:

- Coal-water slurry is prepared at ambient pressure
- Operating temperatures are kept at a minimum of 30°C to allow for use of process cooling water. As a consequence, the liquid contacting is carried out at pressure of 80 bar, rather than 60 bar
- The coal-CO_2 slurry product is concentrated and the slurry is used as gasifier feedstock

3. Methodology

The losses introduced by the CO_2 slurry preparation process had not been accounted for in previous analyses of the performance of CO_2 slurry-fed systems [4]. For the case of the PHICCOS process, these include the coal enthalpy lost through entrainment of coal particles in the ash refuse stream, as well as the power required to recompress CO_2 desorbed in the slurry concentrator and refuse depressurization tanks. PHICCOS, nevertheless, also positively influences the system performance through a reduction of the ash and moisture content of the gasifier feedstock.

Steady-state process simulation is used to quantify the net performance of a coal gasification-based plant with carbon capture and PHICCOS-based coal-CO_2 slurry feed. The application selected is an IGCC power plant. However, this feeding system concept is applicable to any gasification-based plant with carbon capture, including coal-to-liquids facilities.

The economic attractiveness of the PHICCOS feeding system is assessed based on a cost of electricity calculation and through comparison with competing feeding system technologies.

3.1. Process model

An Aspen Plus (A+) model of the PHICCOS process was developed and integrated into a model of a ~600 MW IGCC power plant with 90% overall CO_2 capture and CO_2 slurry feed. The IGCC model has been described elsewhere in detail [4, 13].

The phase inversion step of the PHICCOS process is relatively well characterized empirically thanks to the work conducted during the LICADO project [12]. The operating conditions and expected performance used for the process model were hence adopted from that work, where appropriate. The main PHICCOS modeling assumptions are listed in Table 1.

Table 1. Operating and performance assumptions for PHICCOS model [12]

PHICCOS parameter	Nominal value	Range
Coal-water slurry loading	20 wt.-% ar coal	
Ratio of CO_2 to CWS	0.5 (by weight)	
Liquid contacting vessel		
Operating pressure	80 bar	
Operating temperature	30°C	
Residence time	5 min	
Slurry concentrator pressure	60 bar	
Coal enthalpy recovery	90%	85% - 95%
Coal product composition		
Ash content (dry)	10%	5% - ar
Moisture content	10%	5% - ar

For modeling purposes, the fluid and solid phases were considered separately in the PHICCOS sub-model. The water slurry-CO_2 slurry liquid-liquid equilibrium was modeled as the binary H_2O-CO_2. Minor components in the recirculated CO_2 stream were not considered.

The Predictive Redlich-Kwong-Soave (PSRK) property method was selected for modeling the liquid-liquid phase equilibrium in the water-CO_2 contacting vessel, which is represented by a *DECANTER* model in A+. The PSRK method was found to accurately reproduce the solubility of liquid CO_2 in H_2O, which is has been measured to be 6 wt.-% at 80 bar and 30°C [14] and is thus significant.

The coal stream was modeled separately as a non-conventional component in A+. A fraction of the ash content, moisture, and organic matter in the as-received coal are separated prior to the gasifier to account for the reduction in ash and moisture content in the PHICCOS process, as well as for the coal enthalpy recovery efficiency.

3.2. Cases studied

The cases studied in this work are summarized in Table 2. In order to cover a wide range of coal ranks, both bituminous coal and lignite were considered. The composition of each is presented in the Appendix and corresponds to that used in the National Energy Technology Laboratory's (NETL) Baseline Studies for high and low rank coal [15, 16]. The lignite has a high moisture and ash content of 36% and 15%, respectively, relative to that of bituminous coal with 11% moisture and 11% ash.

Table 2. Cases studied. The asterisk (*) indicates the most economic commercial technology for each feedstock. All cases have a net power output of 500 MW_{el}

Coal	Feeding System	Gasifier
Bituminous	PHICCOS	GE full-quench
	CWS*	GE full-quench
	Dry	Shell
Lignite	PHICCOS	GE full-quench
	CWS	GE full-quench
	Dry*	Shell

The slurry-fed gasifier studied resembles General Electric (GE) technology and has full-quench syngas cooling. This cost-effective cooling method is especially attractive in gasification-based plants with carbon capture, where a high moisture syngas is desirable for the water-gas shift reactor. The latter is particularly true for plants with CO_2 slurry feed, where the low H_2/CO ratio of the syngas requires large amounts of steam in the WGS reactor [4].

State-of-the-art technologies with which the CO_2 slurry feeding system competes were also considered in this work for comparison. The state-of-the-art technologies selected are those which are estimated to yield the lowest cost of electricity in an IGCC plant with carbon capture. For bituminous coal, a GE gasifier with water slurry feed is currently the cheapest option, whereas a Shell gasifier with dry feed is the most attractive one for lignite [15, 16]. These two competing technologies were thus studied for both coals, see Table 2. Unlike all slurry-fed cases, whose performance was estimated with the tools described in this work, the performance of an IGCC plant with a dry-fed Shell gasifier was taken from the literature [15, 16].

The results presented for the PHICCOS feeding system include a realistic estimate, as well as a higher and lower bound, which were defined based on the performance limits observed for the LICADO process. The range considered is defined by a variation of the enthalpy recovery from 85% to 95% and of the product coal ash and moisture content of 5% to its as-received value, see Table 1. Uncertainty bars are used to illustrate the upper and lower limits in the results. Nevertheless, it is unlikely that the process performance will reach the limits because, as the experiments for LICADO showed, the variables considered above are not independent but rather tightly correlated.

3.3. *Economic assessment*

The capital costs for the PHICCOS feeding system were estimated based on Westinghouse's economic assessment of a commercial-scale LICADO unit [12], whereas equipment differences for the two processes were considered. Capacity scaling and cost indices were used, where appropriate. The main economic assumptions used in this work are presented in the Appendix. A process contingency of 40% was used for the proposed PHICCOS process given the uncertainty associated with its costs.

The levelized cost of electricity (LCOE) of the IGCC plants considered is used as the figure of merit for comparing the economic attractiveness of the PHICCOS feed with that of alternative feeding systems. It is a simple way of bringing the performance and cost tradeoffs into a single number and is also a good qualitative indication of the relative merits of individual technologies, even for applications other than IGCC. The cost of electricity model used in this work follows the standard methodology defined by NETL [17]. All bare erected costs for the IGCC process units are also based on NETL's estimates for the same plant and coal type considered here [15, 16]. Operating and maintenance (O&M) costs for a system based on PHICCOS feed are assumed to be the same as those of a coal-water slurry fed plant of comparable size.

4. Results and discussion

The net power generation efficiency of an IGCC plant with carbon capture, a single-stage, full-quench gasifier and coal-CO_2 slurry feed prepared with the PHICCOS process is presented in Figure 3. The resulting cold gas efficiency is also presented. The latter accounts for the losses introduced by the gasifier only and does not include feeding system losses.

The results are compared with that calculated for a similar system based on coal-water slurry. Previous estimates for a coal-CO_2 slurry-fed plant, which did not account for the losses introduced with the slurry preparation, are also presented‡ [4].

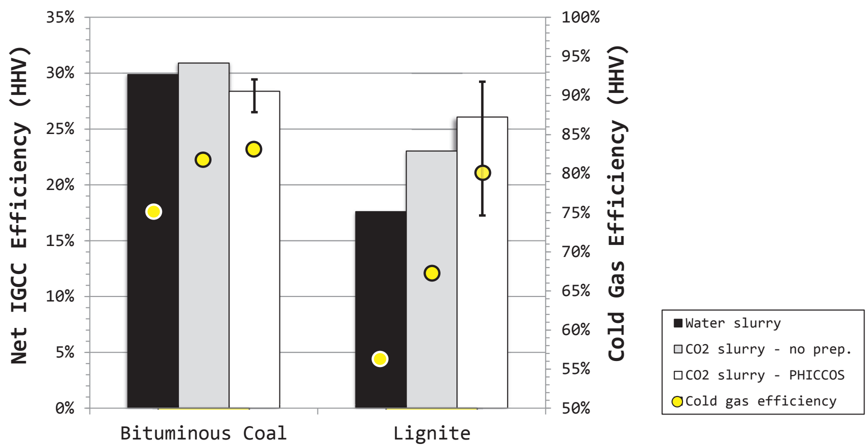

Figure 3: Net IGCC efficiency (left axis) for plant with coal-water slurry and coal-CO_2 slurry without considering slurry preparation [4] and with PHICCOS slurry preparation. The corresponding gasifier cold gas efficiency (right axis) is also shown.

‡ Estimates for lignite have been updated for the different lignite composition used here

The figure shows that the cold gas efficiency for a gasifier with PHICCOS feed is higher than previously estimated for coal-CO_2 slurry without slurry preparation, in particular for the case of lignite. This is a result of the significant coal ash and moisture content reduction expected when coal-CO_2 slurry is prepared through phase inversion, see Table 1.

For a coal with low ash and low moisture like the bituminous coal considered here, the results show that the PHICCOS slurry preparation losses outweigh the modest cold gas efficiency benefits brought by the use of CO_2 as slurrying medium. PHICCOS leads to an almost 2%-point lower efficiency than CWS feed. For lignite, however, the cold gas efficiency of CO_2 slurry gasification is calculated to be much higher, given its high ash and moisture content. The PHICCOS feed hence leads to over 5%-points net IGCC efficiency benefit, relative to coal-water slurry. This performance advantage is, nevertheless, strongly dependent on the PHICCOS process performance, as the upper and lower range represented by the error bars indicates.

Figure 4 presents the bare erected capital costs estimated for the PHICCOS feeding system. The corresponding costs of a coal-water slurry and a dry feeding system with the same coal throughout are also shown for comparison.

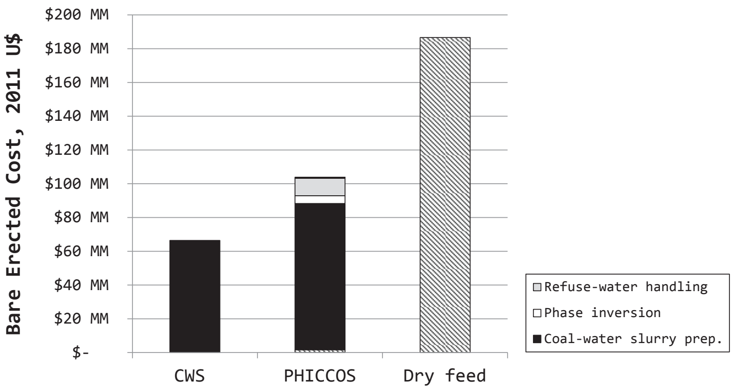

Figure 4: Capital cost of PHICCOS feeding system and comparison with alternative technologies [15, 16]. All costs are for 200 tonne/h as-received coal and include coal handling, preparation and feeding. Process contingency is not included here.

The figure shows that for a given as-received coal flow, the capital costs of the PHICCOS process are about 60% higher than that of coal-water slurry feed, but still only about half of that of a dry feeding system. The additional costs of PHICCOS, relative to CWS, come primarily from the larger coal-water slurry preparation equipment but also from the refuse water handling. The former is estimated to be about five times larger than for conventional coal-water slurry systems given the low CWS loading of 20% in the PHICCOS process.

The pressurized H_2O-CO_2 contacting equipment, where phase inversion occurs, is predicted to have a modest contribution to the capital costs of PHICCOS. It is estimated that 4 vessels of about 60 m^3 each are required to provide the 5 minute total residence time required in the mixing/separating process. While each tank hence has approximately the same volume as the reaction section of the gasifier, it is relatively standard equipment operating at ambient temperature so despite the large size, its capital costs are expected to be low compared to other, more complex process units.

Finally, Figure 5 presents the levelized cost of electricity estimated for an IGCC plant with 90% CO_2 capture and PHICCOS-based coal-CO_2 slurry feed. The LCOE for a plant based on conventional coal-water slurry feed and dry feed is also shown for comparison. All IGCC cases have a net output of 500 MW_{el}.

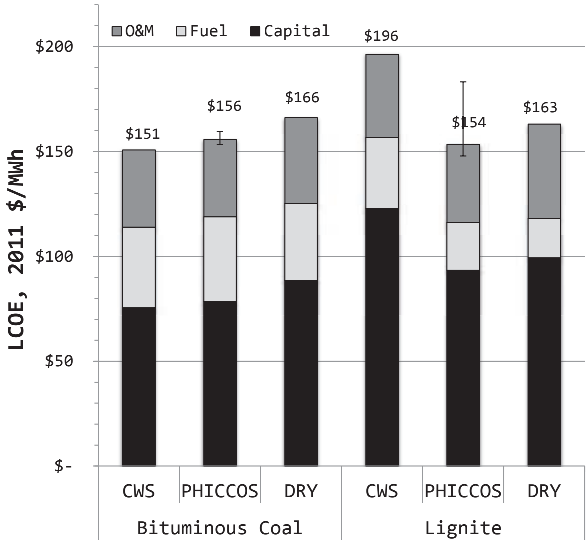

Figure 5: Levelized cost of electricity comparison for 500 MW_{el} IGCC plant with CCS and different feeding systems

For bituminous coal, the results show that a plant with PHICCOS produces electricity at $156/MWh, which is somewhat more expensive than conventional coal-water slurry feed at $151/MWh. This is expected since the efficiency is lower and the capital costs higher than for CWS, see Figure 3 and Figure 4. Nonetheless, the PHICCOS feeding system is significantly more attractive than the capital-intensive dry feed with an LCOE of $166/MWh.

For lignite, on the other hand, a plant with PHICCOS produces electricity at $154/MWh and hence more cost effectively than the competing technology, which for low-rank coal is the dry feeding with an LCOE of $163/MWh. In this case, coal-water slurry feed is very expensive at $184/MWh, despite its low capital costs, as a result of the very low efficiency of low-rank coal-water slurry gasification.

The error bars in Figure 5 illustrate the importance of the phase inversion performance on the PHICCOS feeding system economics for the case of lignite. It is important to note, however, that the upper limit shown in the figure merely defines the maximum possible LCOE for PHICCOS. The conditions required to reach that limit (no ash reduction, no moisture reduction, low enthalpy recovery), however, are unlikely to occur simultaneously because these variables are physically coupled. Further experimental work is required for a better characterization of the performance envelope.

5. Conclusions

The Phase Inversion-based Coal-CO_2 Slurry (PHICCOS) process is proposed in this work as a means to prepare coal-CO_2 slurry at ambient temperature for feeding pressurized, single-stage, entrained flow gasifiers. Steady-state process simulation was used to quantify the performance of a gasification-based plant with the PHICCOS feeding system, whereas an IGCC plant with carbon capture was used as an example application. Experimental results for coal-water slurry inversion were used as a basis to design the process and construct the process model.

The PHICCOS preparation and feeding system was found to influence the plant performance in different ways; it is detrimental through the coal enthalpy lost during phase inversion as well as through even higher CO_2 recompression requirements than previously estimated, due to the high solubility of CO_2

in water. It is beneficial through a reduction of ash and moisture content of the feedstock. The net efficiency benefit, relative to coal-water slurry, depends on the characteristics of the coal in question.

For bituminous coal, an IGCC plant with PHICCOS feed was found to be about 2%-points less efficient than a coal-water slurry fed plant. In the case of lignite feedstock, PHICCOS feeding leads to a 5%-point higher net efficiency. Nonetheless, the capital costs of PHICCOS are about 60% higher than for CWS.

Overall, the levelized cost of electricity is estimated to be \$156/MWh for bituminous coal, which is only marginally higher than the competing technology, which is coal-water slurry feed. For lignite, the calculated \$154/MWh is lower than for the competing dry-fed system.

Overall, the PHICCOS feeding system offers an attractive tradeoff between efficiency and capital investment. The consistently low ash and moisture content of the gasifier feedstock it produces has the potential to significantly increase the feedstock flexibility of gasification-based plants. Unlike coal-water slurry or dry feeding systems, which are attractive only for high or low-rank coal, respectively, PHICCOS is cost-effective across the entire coal rank spectrum and is particularly appealing for the economic utilization of coal with high moisture and/or high ash content.

6. Outlook

Future work should focus on gaining a better understanding of the mechanism underlying the phase inversion of coal by CO_2, which is known to be surface-property driven but is not yet fully understood [18].

The effect of the operating conditions on the phase inversion process performance should also be studied in more detail. It is important to understand which conditions are desirable for operating the PHICCOS process, which, as opposed to LICADO, is not targeting coal beneficiation but merely benefits from it. The phase inversion behavior of low-rank coal is of especial relevance, as it is known to be less hydrophobic than high-rank coal. This could require the addition of surfactants to the PHICCOS process, which have shown to be effective at increasing the hydrophobicity of the coal surface.

Finally, the impact of the feedstock ash content on the gasifier performance and operability should be studied, including its effect on cold gas efficiency, slag removal system, and on the protective slag layer on the gasifier wall.

Acknowledgements

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Appendix

Table 3: Proximate analyses of coals considered (dry basis)

	Bituminous [15]	Lignite [16]
Moisture (ar)	11.12%	36.08%
Ash	10.91%	15.43%
Volatile Matter	39.37%	41.49%
Fixed Carbon	49.72%	43.09%
Higher Heating Value	30,506 kJ/kg	24,254 kJ/kg

Table 4. Main economic assumptions

Parameter	Value/Source
Cost basis	2011
EPC Costs	0.09 of bare erected cost
Owners costs	0.23 of total plant cost
IGCC Capital and O&M Cost	[15, 16]
PHICCOS capital cost basis	[12]
Capital Charge Factor	0.1243
Bituminous coal price	$49.46/ton [19]
Lignite price	$14.57/ton [19]
Scaling exponent	0.6
Cost Index	Chemical Engineering Plant Cost Index
Capacity Factor	0.8
Levelization Factor	1.268
Project Contingency	14%
IGCC Process Contingency	5%
PHICCOS Process Contingency	40%